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Neutron Scattering Group

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Research Activities

A highlight in our group was that Masahumi Kohgi was promoted to the professor of the Tokyo Metropolitan University, and he moved there in January. He served for more than 20 years in Tohoku University and he created an important research field of the magnetism of the rare earth metals, in particular his recent works on the neutron studies from CeP and related compounds are internationally recognized. He is busy now in establishing his research group there. Kenji Nakajima who has been active as the JSPS Research Fellow for two years in our group was promoted to the Research Scientist at ISSP, University of Tokyo. Kazuma Hirota joins us in our group on April this year as the new Research Scientist. He has been working at the Physics department, Brookhaven

National Laboratory. He was recognized with his recent works on two-length scale in magnetic critical scattering studies. Since he has an excellent skill in the diffraction experiments, we expect his important role in our group in research and teaching.

Another highlight was that the International Conference on Neutron Scattering (ICNS' 94) (Y. Endo was the chairman of the conference organizing committee) was held on October 11th-14th, the last year at the Sendai International Center. The Sendai conference is the first of a series of the international conference on the neutron scattering research in condensed matter science. It was an unexpected coincidence that the 1995 Nobel prize in the physics was going to the pioneers of the neutron scattering and diffraction, Bert Brockhouse (McMaster University) and Cliff Shull (M.I.T.). About 400 participants from all over the world celebrated them at the conference.

We have gained a very important step in the research of the high-temperature superconductivity by observing a first evidence of the gap in the magnetic excitations in the superconducting state of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ ($T_c=37.3\text{K}$). Our research including the international collaborations such as Tohoku - MIT - BNL, Tohoku - HMI, and Tohoku - Kobe - DRAL are going very smoothly. We have continuously produced a number of important outcomes including the high-temperature superconductivity, which will be briefly described in each section of our research projects. The installation of the single crystal growth as well as the measurements is also going as we aimed under the support of the research projects. For instance, a new high-precision SQUID susceptometer, a superconducting magnet for neutron scattering, a special furnace for the sample heat treatment and a new closed cycle cryostat will arrive in this semester.

(I) High temperature superconductivity and related subjects

a) Single crystal growth and preparation

(N. Kaneko, C.H. Lee, S. Wakimoto and J. Wada)

We have grown single crystals of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ with various Sr concentrations. Since the growth-technique for $x=0$ and 0.15 has been established, we have been developing the technique to grow crystals with the larger diameter and to control the grown crystal orientation, both of which are quite important for the quantitative neutron scattering experiment. For the under doped region ($x<0.15$), $x=0.10$ crystal ($T_c=25.5\text{K}$) with the volume of about 0.2cc and for the over doped region, $x=0.2$ and 0.18 crystals ($T_c=25.5\text{K}$ and 29K , respectively) with the volume of about 0.1cc were grown by using TSFZ (Traveling Solvent Floating Zone) method. Since there exists no sizable single crystal of $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$, one of the ideal electron-doped high T_c superconductors for the neutron scattering experiment, we started to grow the single crystal by TSFZ method. After searching for the

growth condition, several sizable grains of the single crystal were obtained.

Parallel to such single crystal growth, the "oxygen controlled" samples of $\text{La}_2\text{CuO}_{4+d}$, $\text{La}_2\text{NiO}_{4+d}$ and $(\text{La}(\text{Ba}))_2\text{CuO}_{4+d}$ were prepared by the post-growth heat treatment.

This subject is collaborated with Yamanashi University (S.Hosoya).

b) Spin fluctuations in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

(K.Yamada, Y.Endoh and C.H.Lee)

Neutron inelastic scattering experiments have been performed on our single crystal of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ with the highest T_c (37.3 K at onset) among the previously studied single crystals. The thermal evolution of the low energy incommensurate peak intensity near (p,p) exhibits a pronounced peak at around 40 K. In contrast to the results on the previous crystals, the intensity below about 3.5 meV dramatically decreases as the temperature decreases below T_c and eventually is vanished into the background below around 15 K. The energy spectrum of the intensity at low temperature also exhibits a gap like behavior below about 3.5 meV. The observed size of the energy-gap is much smaller than the superconducting gap of the BCS theory suggesting the anisotropic d-wave symmetry for the pairing state in this system.

This subject has been collaborated with Brookhaven National Laboratory (G.Shirane), Massachusetts Institute for Technology (R.J.Bergeneau, M.A.Kastner) and Yamanashi University (S.Hosoya).

c) Superconductivity, phase separation and spin correlation in $(\text{La},\text{Ba})_2\text{CuO}_{4+d}$

(S.Wakimoto, K.Yamada and Y.Endoh)

The role of interstitial excess oxygen atoms in the high- T_c cuprates is of interest to study because the induced carriers dramatically vary the magnetic as well as superconducting properties of these oxides. In $\text{La}_2\text{CuO}_{4+d}$, two-phase separation in the polycrystalline sample was reported by structural analyses. We performed systematic heat treatments using single crystals of $\text{La}_{2-x}\text{Bi}_x\text{CuO}_{4+d}$ to study the effect of oxygen doping on the phase stability as well as the three-dimensional magnetic order. For $\text{La}_2\text{CuO}_{4+d}$, the analysis of magnetic susceptibility peak near the Ntemperature T_N revealed that the continuous decrease of T_N upon doping is terminated at 280 K. The further doping additionally induces the previously reported macroscopic phase separation accompanying an antiferromagnetic phase with $T_N=265$ K and a superconducting one with $T_c \sim 35$ K. On the other hand, for $\text{La}_{2-x}\text{Bi}_x\text{CuO}_{4+d}$ with $x \sim 0.05$ no macroscopic phase separation was

observed with d up to around 0.02. Upon the continuous oxygen doping, the T_N continuously decreases from 310 K to below 10 K only by the heat treatment. The scaling behavior of magnetic susceptibility previously studied by the powder sample of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4+d}$ was reexamined and found out a deviation from the scaling law.

d) Lattice vibrations in high- T_c superconductor

(K.Yamada and L.H.Lee)

Neutron inelastic scattering measurements on the single crystals of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_{4-y}$ have been performed to study the relation between the soft phonon and the high- T_c superconductivity. The phonon softening of the Z-point mode due to the incipient structural phase transition to the low temperature tetragonal ($P4_2/nm$) or orthorhombic ($Pccn$) phase was appeared to be frozen below T_c . The narrowing of the phonon line-width upon cooling was saturated below T_c , suggesting the appearance of the extra line broadening in the superconducting state.

This work was done in collaboration with Kobe University (M.Arai, K.Ubukata and M.Fujita)

(II) Magnetism in the transition metals

a) Metallic ferromagnetism in CoS_2

(H.Hiraka, Y.Endoh and K.Yamada)

CoS_2 is a ferromagnet exhibiting a typical itinerant character, which stimulates a continuous interest for more than decades. We recently showed the experimental evidence that the magnetic phase transition is really of the second order but presumably near the tricritical point suggested by the unusually large values of the critical indices α and α' . Spin dynamics are now investigated by using neutron scattering techniques from assebled single crystals. At this moment, we found very isotropic spin wave mode at lowest temperature. Now we extend our studies to the higher temperatures across the Curie temperature.

On the oher side, the single crystals of $\text{Co}(\text{S}_{1-x}\text{Se}_x)_2$ have been grown. The purpose of the studies is to find the tricritical point, which is now going on.

b) Spin excitations in the spin density wave state in Cr

(T.Fukuda, Y.Endoh, M.Takeda and K.Yamada)

Spin excitations in the spin density wave state in Cr have been studied extensively. This study includes not only the scientifically important but technically interest problems due to the extremely

large linear spin wave dispersion relation with respect to the wave vector q . The spin wave velocity is so large that the scattering intensity at the smallest q behaves the proportionality in ω in spite of the ω^{-1} in the usual antiferromagnet.

Nevertheless our experiment clearly shows a three peaked structure in q as well as the anomalous ω dependence in $\chi''(q, \omega)$, which triggers also the theoretical interest. According to the latest calculation in terms of the random phase approximation based on the three band model (R.Fishmann and S.H.Liu), not generally believed two band model, the three peaked structure in q observed in our measurement is quite possible. However the quantitative agreement is not perfect, which will be studied in the future. The energy dependence of $\chi''(q, \omega)$ is still working in order to obtain a reliable result.

c) Metal and insulator transition in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$

(Y.Endoh and K.Hirota)

The perovskite Mn oxides become again a fashionable materials since Tokura found the anomalously giant magnetoresistance (GMR) in this system where $x \approx 0.2$. This anomalous GMR seems to be associated with the metal-insulator transition and the structural transition between an orthorhombic and a rhombohedral structure near this critical x value. The metallic transition was found to be associated with the antiferromagnetic ($x < 0.2$) to ferromagnetic ($x > 0.2$) state. This transition is well known to be a double exchange mechanism but the both the spin dynamical and lattice dynamical studies are very few. We have initiated the studies of magnetism as well as the crystal structure of $x=0.3$ crystal by using neutrons. So far we have observed a quite interesting result of the typical signature of the itinerant ferromagnetism and the crystal distortion resulted by the rotation of the octahedron of oxygen atoms. The predicted distortion at the ferromagnetic transition was not observed.

This study is collaborated with Tokura's group at JRCAT, University of Tokyo and Shirane's group at BNL.

(III) Polarized neutron studies and multilayered superlattices

a) Polarized neutron studies

(M. Takeda, K. Kurahashi, T. Fukuda, M. Onodera and Y. Endoh)

In this fiscal year, we have been improving both polarized neutron spectrometer, TOP, at

KENS and polarized neutron option of TOPAN at JAERI.

TOP has been upgraded with a V-shaped polarizing neutron guide (PNG) last year. As the next step, we redesigned the layout of TOP in order to get more intensity of neutron at sample position with the PNG. Two kinds of experiments were done on TOP spectrometer up to now. One is the experiments with high resolution requirement such as reflectivity and small angle polarized neutron scattering measurements (SANS) and the other is the neutron transmission measurement (depolarization measurement). Although the previous TOP was designed for the small angle neutron scattering machine, these two experiments were done with the same set up. TOP was partly remake to separate and to optimize these experiment, respectively.

The flight path length, L1, which is the distance between the exit of the PNG and the sample position, for depolarization measurement was reduced from 2 m to 0.5 m to avoid the loss of intensity of the incident neutron by the divergence of neutrons. New sample goniometer and a spin flipper of Mezei type were made for this reduction. For depolarization measurements CoTi analyser with single detector is used just after the sample position.

The L1 is also reduced from 2 m to 1 m in the reflectivity and SANS measurements set up. The position sensitive detectors and a detector chamber, which are used for the previous TOP, are used for this set up. The set up for each experiment is easily and quickly switched each other.

The assembly of the new TOP spectrometer has been finished, and now we are making adjustment of each components and preparing for experiments. These work are collaboration with Dr. S. Itoh at KENS.

We have constructed the polarized neutron option of TOPAN spectrometer using Heusler polarizing monochromator crystals and done several polarized neutron scattering experiments. Recently we got the news that a heat treatment significantly improve the performance of polarizing efficiency of Heusler crystals from Dr. Magerl in ILL. We visited him and discussed what is the best way and how to done the heat treatment to get best performance of Heusler crystals. We just start experiments to elucidate the relation between the heat treatment and the polarizing efficiency of the crystals.

This work was performed in collaboration with Dr. Magerl in ILL.

b) Interfacial roughness and the giant magnetoresistance effect of Fe/Cr multilayers

(M. Takeda and Y. Endoh)

Fe/Cr multilayers show a huge negative magnetoresistance and the phenomena is known to be a giant magnetoresistance (GMR) effect. A lot of theoretical and experimental work have been done to clarify to the origin of the GMR effect. Although it is proved that the antiferromagnetic coupling of ferromagnetic moments of Fe layers through the Cr spacer layers are essential to the GMR effect, there is still an open question whether the interfacial roughness between Fe and Cr layers enhances or reduces the GMR effect.

We have grown single crystal Fe/Cr multilayers with different interfacial roughness by molecular beam epitaxy. One is the $[\text{Fe}(3.0 \text{ nm})/\text{Cr}(1.0 \text{ nm})]_{30}$ on MgO and the other is that on sapphire. The neutron experiments were done on TOP spectrometer at KENS and TOPAN at JAERI. The Fe/Cr on sapphire showed the larger GMR effect than that on MgO. The $\theta - 2\theta$ could not make the difference between these two samples clear. The interfacial roughness clearly appears in the off specular diffuse scattering profile. In both samples off specular diffuse scatterings were observed around the antiferromagnetic Bragg peak of the Fe/Cr bilayer. The diffuse scattering of the sample on sapphire more intensively appeared than in that on MgO. This observation clearly indicates that the interfacial roughness enhances the GMR effect. However, it is important to estimate the interfacial roughness quantitatively because it is clear that the extremely large roughness reduces the GMR effect.

This work was performed in collaboration with Dr. Mizuki and Dr. Kamijo in NEC corp.

Publications

- (1) *Local Structural Instability of High- T_c Oxide Superconductors Studied by Inelastic Neutron Scattering*, M. Arai, K. Yamada, S. Hosoya, A.C.Hannon, Y. Hidaka, A.D. Taylor and Y. Endoh, *Journal of Superconductivity* **7** (1994) 415-418.
- (2) *Magnetic Excitation in Spin Density Wave (SDW) State of Cr*, Y. Endoh, T. Fukuda, K. Yamada and M. Takeda, *J. Phys.Soc.Japan*, **63** (1994) 3572-3576.
- (3) *Spin Correlations in the 2D Heisenberg Antiferromagnet $\text{Sr}_2\text{CuO}_2\text{Cl}_2$: Neutron Scattering, Monte Carlo Simulation, and Theory*, M. Greven, R. J. Birgeneau, Y. Endoh, M. A. Kastner, B. Keimer, M. Matsuda, G. Shirane and T. R. Thurston, *Physical Review Letters* **72** (1994) 1096-1099.
- (4) *Spin correlations in $\text{Sr}_2\text{CuO}_2\text{Cl}_2$* , M. Greven, R. J. Birgeneau, Y. Endoh, M. A. Kastner, B. Keimer, M. Matsuda, G. Shirane and T. R. Thurston, *Physica B* **199 & 200** (1994) 642-

643.

- (5) *Ferromagnetic Transition of Heisenberg Ferromagnetic Metal of CoS_2 ---Static Critical Properties---*, H. Hiraka and Y. Endoh, J. Phys. Soc. Japan, **63** (1994) 4573-4582.
- (6) *Single Crystal Growth of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ with Improved Lamp-Image Floating-Zone Furnace*, S. Hosoya, C. H. Lee, S. Wakimoto, K. Yamada and Y. Endoh, Physica C**235-240** (1994) 547-548.
- (7) *Quantum Spin Dynamics of an S-3/2 Heisenberg Antiferromagnetic Chain-Inelastic Pulsed Neutron Scattering from CsVCl_3* , S. Itoh, Y. Endoh, K. Kakurai and H. Tanaka, Phys. Rev. Lett.**74** (1994) 2375-2378.
- (8) *Spin Dynamics on Two-Dimensional Heisenberg Antiferromagnets --- High Energy Neutron Inelastic Scattering from La_2CuO_4 and La_2NiO_4 ---*, S. Itoh, K. Yamada, M. Arai, Y. Endoh, Y. Hidaka and S. Hosoya, J. Phys. Soc. Jpn. **63** (1994) 4542-4549.
- (9) *Single Crystal Neutron Diffraction Study of the Magnetic Phase Diagram of the Heavy Fermion Superconductor UPd_2Al_3* , H. Kita, A. Donni, Y. Endoh, K. Kakurai, N. Sato and T. Komatsubara, J. Phys. Soc. Jpn. **63** (1994) 726-735.
- (10) *Spin fluctuation in superconducting $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$* , M. Matsuda, K. Yamada, Y. Endoh, T. R. Thurston, G. Shirane, R. J. Birgeneau, M. A. Kastner, I. Tanaka and H. Kojima, Physical Review B**49** (1994) 6958-6966.
- (11) *Spin fluctuations in insulating, weakly metallic and superconducting $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$* , G. Shirane, R. J. Birgeneau, Y. Endoh and M. A. Kastner, Physica B**197** (1994) 158-174.
- (12) *Weak Ferromagnetism and Antiferromagnetic Ordering of 2p Electrons in Organic Radical Compound 2,4,6-Triphenylterdazyl*, S. Tomiyoshi, T. Yano, N. Azuma, M. Shoga, K. Yamada and J. Yamauchi, Phys. Rev. B**49** (1994) 16031-16034.
- (13) *Preparation of single crystal of Ca_2CuO_3 by TSFZ method*, J. Wada, S. Wakimoto, S. Hosoya, K. Yamada and Y. Endoh, Physics C**244** (1995) 193-197.

Doctor Thesis

D1) Neutron scattering study on the low carrier materials (T. Osakabe)

Master Thesis

- M1) Single crystal growth and characterization of the superconducting cuprates (S. Wakimoto)
- M2) A study of the superconducting flux state using polarized neutron (K. Kurahashi)
- M3) Neutron scattering study at milli-Kelvin region :Study of heavy Fermion magnets: (H. Okuyama)